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# Bi-ferroic memristive properties of multiferroic tunnel junctions

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The giant tunnelling electroresistance (TER) and memristive behaviours of ferroelectric tunnel junctions make them promising candidates for future information storage technology. Using conducting ferromagnetic layers as electrodes results in multiferroic tunnel junctions (MFTJs) which show spin dependent transport. The tunnelling magnetoresistance (TMR) of such structures can be reversibly controlled by electric pulsing owing to ferroelectric polarisation-dependent spin polarisation at the ferroelectric/ferromagnetic interface. Here, we show multilevel electric control of both TMR and TER of MFTJs, which indicates the bi-ferroic or magneto-electric memristive properties. This effect is realised by manipulating the ferroelectric domain configuration via non-volatile partial ferroelectric switching obtained by applying low voltage pulses to the junction. Through electrically modulating the ratio between up- and down-polarised ferroelectric domains, a broad range of TMR (between ~3% and ~30%) and TER (~1000%) values can be achieved. The multilevel control of TMR and TER using the electric pulse tunable ferroelectric domain configuration suggests a viable way to obtain multiple state memory. *Published by AIP Publishing.* https://doi.org/10.1063/1.5023877

Magnetic tunnel junctions (MTJs), composed of a thin dielectric barrier sandwiched between two ferromagnetic electrodes, present the tunnelling magnetoresistance (TMR) effect which shows two distinct resistance states in response to the parallel or antiparallel magnetic moment configuration in the ferromagnetic electrodes. Although MTJs have been the subject of intensive study due to their great potential for non-volatile magnetic memory applications,[1–3] moderate data storage density and especially a high-power consumption for controlling magnetisation still remain major challenges. To tackle those problems, several methods have been proposed including but not limited to utilising various non-collinear magnetization configurations in the ferromagnetic electrodes of semiconductor spin valves[4] or all-oxide MTJs,[5] manipulating domain states of ferromagnetic electrodes in pseudo spin valves,[6] or tuning tunnel barrier oxygen vacancies by voltage pulses in MgO-based MTJs.[7]

Despite the aforementioned solutions, one promising candidate is multiferroic tunnel junctions (MFTJs), in which the ferroelectric/multiferroic layer is incorporated into a conventional magnetic tunnel junction (MTJ) as the tunnel barrier.[8–16] Four memory states can be obtained in MFTJs by voltage pulsing thanks to the coexisting tunnelling magnetoresistance (TMR) and tunnelling electroresistance (TER) given by resistance variation upon different ferroelectric polarisation orientations.[17–24] Moreover, MFTJs have been demonstrated as a rich platform to explore the electric control of magnetism at the ferroelectric/ferromagnetic interface,[9,11] which makes such a device appealing for high density memory applications with ultralow power consumption and from the research point of view.

As mentioned before, four resistance states can be obtained for an MFTJ upon switching the ferroelectric single-domain state. However, during the ferroelectric switching, domains with opposite polarisations can indefinitely coexist. The actual domain configuration can be easily tuned by choosing the parameters, i.e., amplitude and duration, of the electric switching pulse.[25] The domain configuration, an important degree of freedom in ferroelectrics, can in principle result in multiple levels of ferroelectric polarisation states corresponding to the volume ratio of up- and down-polarised ferroelectric domains.[26–28] Recently, engineering of the ferroelectric domain structure has enabled new functionalities in ferroelectric-based devices, such as multilevel polarisation states in the ferroelectric random-access-memory (FeRAM)[27] and continuous resistance states in the ferroelectric memristor.[26,28,29] On the other hand, it has been shown that in an MFTJ, significant change of spin-dependent tunnelling and thus TMR can be obtained between opposite ferroelectric polarisation states due to the polarisation dependent screening effects[30] and hybridisation[10,13] at the ferroelectric/ferromagnetic interface. Keeping this in perspective, like the case of memristor behaviour in ferroelectric tunnel junctions (FTJs), one can perceive that the spin transport, respectively, the value of TMR, in an MFTJ can be continuously tailored by controlling the ferroelectric domain configuration. So far, this effect of the ferroelectric domain configuration on MFTJ magnetotransport has not been investigated.

In this report, we experimentally demonstrate the multilevel electric control of both TMR and TER in Co/$PbTiO_3$(PTO)/$La_{0.3}Sr_{0.7}MnO_3$(LSMO) MFTJs. While in the case of ferroelectric tunnel junctions, the ferroelectric polarisation tunes the charge transport (tunnel current), in the present case, the ferroelectric polarisation controls the spin polarised transport and thus TMR of MTJs. This electric field multi-level control of TMR and TER shown here suggests electric and magnetic multiple memory levels or bi-ferroic memristive nature as an enhanced functionality of MFTJs.

MFTJs are based on high quality PTO (12 u.c.)/LSMO (60 u.c.) epitaxial thin films grown on $SrTiO_3$ (STO) substrates by Pulsed Laser Deposition (PLD). The detailed

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fabrication process can be found elsewhere.[14] High quality PTO/LSMO films are characterised by high resolution scanning transmission electron microscopy (STEM) and atomic force microscopy (AFM) (see Fig. 1). AFM and piezoresponse force microscopy (PFM) images were collected using the Park XE-100 system. PFM data were collected with an ac voltage of 1 V and a frequency of 23.27 kHz using a NSC14/Pt (MikroMasch) cantilever. TEM images were taken using a double CEOS corrected (to third order), Schottky emission JEOL ARM-200F microscope operating at 200 kV in the STEM mode. TER and TMR of MFTJs were characterised using a Keithley 6517 electrometer and a Tektronix AFG 3102 waveform generator in a commercial Physical Properties Measurement System (PPMS) with the temperature range from 2 K to 400 K and magnetic field up to 9 T. All TMR measurements were conducted at 10 K after cooling the sample from 300 K with $-0.5$ T.

Atomic number contrast in annular dark field (ADF) STEM imaging shows that the LSMO/PTO interfaces are atomically sharp, as shown in Fig. 1(a). It can also be seen from the AFM image in Fig. 1(b) that the surface of the PTO is atomically flat. The ferroelectricity of the PTO film is shown in Fig. 1(c); the out-of-plane PFM image demonstrates clear 180° phase contrast after electric writing of ferroelectric domains, which indicates strong piezoelectric activity and stability of written ferroelectric domains of our thin PTO films even at 12 unit cell thickness. Finally, the tunnel junction devices are defined depositing Au/CoO/Co top layers and pattern them ($40 \times 40$ $\mu m^2$) as top electrodes by photolithography, sputtering, and lift-off.

The main functional properties of the tunnel junction are shown in Fig. 2. The TER properties are shown in Fig. 2(a) where zero-magnetic field *I-V* curves of the MFTJ were collected at 10 K after applying writing voltage pulses with various amplitudes. Starting from the totally downward polarised state (after 6 V poling), we gradually changed the population of upward and downward ferroelectric polarisation via a set of negative voltage pulses. The transport properties of our MFTJ change accordingly, and eventually, the full change of the resistance state associated with the single domain state (*P* fully upwards) has been obtained. Summarised in Fig. 2(b),

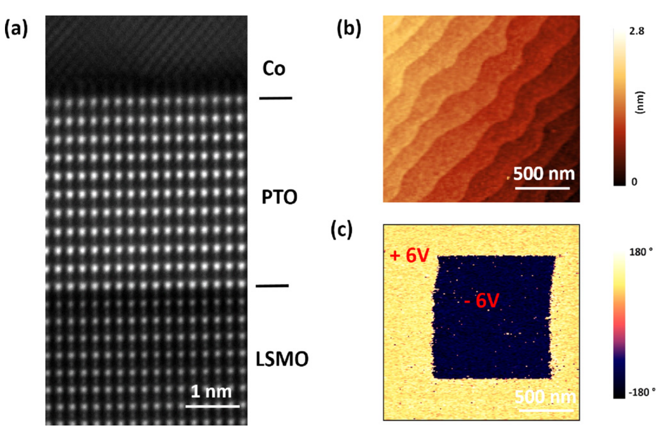


FIG. 1. (a) ADF STEM image of the Co/PTO (12 u.c.)/LSMO heterostructure where sharp interfaces are observed. (b) AFM image of the PTO/LSMO film showing an atomically flat surface. (c) Out-of-plane PFM phase image, the contrast shows the ferroelectric polarisation states after 6 V and $-6$ V dc bias writing.

one can clearly see this reversible resistive switching behaviour of our MFTJ. The multilevel resistance states change shown here, demonstrating the memristive properties, can be attributed to the corresponding domain reconfiguration process during polarisation reversal in the ferroelectric films similar to previous reports.[26,29,31,32]

To gain insights into the MFTJ spin transport properties in response to the ferroelectric barrier domain configuration, we performed typical magnetotransport measurements after cooling the sample at $-0.5$ T to 10 K from room temperature. Figures 2(c) and 2(d) show the continuous and remnant tuning of TMR with applied external voltage pulses to switch partially the polarisation. The TMR is defined as: $TMR = (R_{ap} - R_p)/R_{ap}$, where $R_{ap}$ and $R_p$ are the junction resistance in the antiparallel and parallel magnetic configurations of the electrodes, respectively. The negative TMR observed here suggests a negative spin polarisation nature at the Co/PTO interface and highlights the spin-dependent interfacial bonding effect typical to the perovskite oxide-based interface.[10,11,14,33] Note also the asymmetry of TMR which most probably is due to the exchange bias effect of Co/CoO after field cooling where the native CoO induces a larger ferromagnetic coercivity.

TMR is found to strongly depend on the ferroelectric polarisation orientation. More than 10-fold change of the TMR value is observed here between up- and down-polarised ferroelectric single domain states. Unlike in our earlier work on $Pb(Zr,Ti)O_3$-based tunnel junctions, the TMR does not change the sign as the polarisation switches from totally up ($P_{up}$) to totally down ($P_{down}$) states.[9,14] In the present case of PTO-based tunnel junctions, the TMR absolute value decreases from about 30% in the $P_{down}$ state to almost zero, i.e., only 3%, in the $P_{up}$ state, rather similar to the $BaTiO_3$ (BTO) case reported by Garcia *et al.*[11] It is worth noting that the variation of TMR amplitude with the PTO polarisation direction is opposite to that of the BTO case, i.e., TMR absolute value gets larger while switching PTO polarisation downward which is consistent with previous Pb-based ferroelectric MFTJs.[14,34]

Given the ability to partially switch the polarisation, in other words, to continuously modify the population of $P_{up}$ and $P_{down}$ ferroelectric domains, we can assume that the TMR will follow the same tendency as the TER. In such a way, an intrinsic spin transport property of the junction, i.e., TMR, can be continuously tuned by solely applying the electric field pulses. Since the tunnel transport related to both magnetic and electric order parameters is electrically tuneable, we can state that MFTJs are bi-ferroic or magnetoelectric memristors. To confirm that, we switch stepwise the ferroelectric polarisation of the MFTJ by applying consecutive voltage pulses with various amplitudes and a fixed duration of 50 $\mu$s at zero magnetic field. As expected, a clear stepwise TMR evolution from a high value (31%) towards a low value (2.8%) in response to the ratio of up- and down-polarised ferroelectric domains is observed, as shown in Fig. 2(c). TMR changes more than an order of magnitude, or more than 1000% change in terms of the $TMR_{high}/TMR_{low}$ ratio, by applying voltage pulses between $-6$ V and 6 V. This effect which leads to a multi-level electric control of TMR is reversible and reproducible. As can be seen in Fig. 2(d), multiple TMR levels between high ($\sim$31%) and low

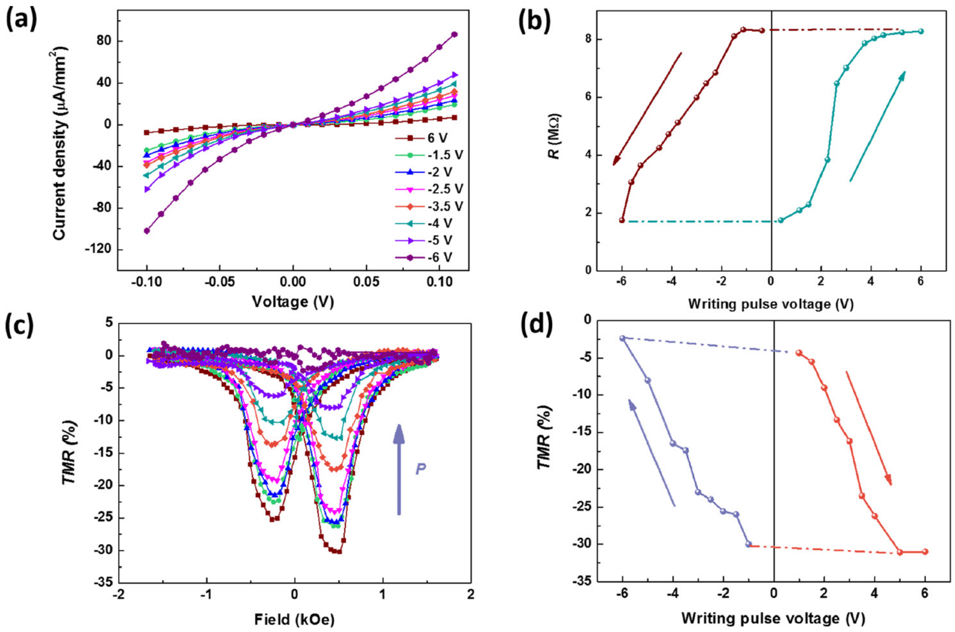


FIG. 2. (a) $I-V$ curves of the MFTJ measured at 10 K after poling by writing voltages pulses with different amplitudes and a constant width (50 $\mu$s). (b) Tunnel resistance of the MFTJ after poling the PTO ferroelectric barrier into different states. (c) TMR of the junction recorded at 10 K with 10 mV applied bias after −0.5 T cooling from 300 K as a function of PTO ferroelectric polarisation state. (d) Dependence of the TMR of the PTO polarisation states as changed by various writing pulse voltages.

($\sim$2.8%) states can be deterministically set in a classical hysteretic way that follows ferroelectric polarisation switching. These results unambiguously demonstrate that depending on the ferroelectric polarisation switching history, the TMR states of the MFTJ can be finely tuned in any level between $TMR_{high}$ and $TMR_{low}$.

Next, we show that the multilevel control of TMR is not only dependent on the voltage pulse amplitude but also on the duration which provides a further parameter for control from the application point of view. As shown in Fig. 3, the TMR changes as a function of voltage pulse duration under 5 V and −6 V. The measurement sequence is shown in Fig. 3(a). We performed each TMR measurement after applying a SET pulse with different durations to the junction. Before each SET pulse and TMR measurement, we reset the junction into the $TMR_{high}$ or $TMR_{low}$ state as the reference by applying a 10 ms-long −6 V or 5 V ReSET pulse. The results are summarised in Fig. 3(b). Clearly, starting from the reference state, the TMR can be driven into a large number of intermediate states between $TMR_{high}$ and $TMR_{low}$ using certain writing pulses. The time scale to access those intermediate levels is of $\sim\mu$s range which is comparable with the ferroelectric polarisation switching time.[26,28] Overall, the results shown in Fig. 3 demonstrate that the TMR levels can be tuned by not only the voltage pulse amplitude but also the period of the switching pulse. This is in agreement with the ferroelectric polarisation switching kinetics[28,35] and further confirms that the multiple TMR states correspond to the ferroelectric barrier domain configuration.

The microscopic mechanism behind the multi-level control of TMR in our MFTJs is related to the ferroelectric polarisation dependent spin transport and multi-domain state of the tunnel barrier. The ferroelectric polarisation switching process, which involves the domain nucleation and growth, can result in a stable domain pattern of mixed domains with opposite polarisations.[26,28] A schematic of such a mixed domain state is sketched in Fig. 4(a) (see also Fig. S3 for the PTO ferroelectric domain evolution recorded by PFM). Through carefully choosing the switching pulse amplitude and time, one can obtain exactly the desired ratio of the up- and down-polarised domains.[26–29] Moreover, the multi-domain state is robust due to its smaller depolarisation energy than that of the single-domain state.[25]

Although the mechanism that relates the spin transport and ferroelectric polarisation remains under debate, a plausible scenario suggested by recent theoretical and experimental work is related to emerging hybrid electronic and magnetic states or even ultrathin interfacial metal oxide layers which can form at the ferromagnetic/ferroelectric interface and be manipulated by ferroelectric polarisation.[9–11,14,33] This interface acts as a further

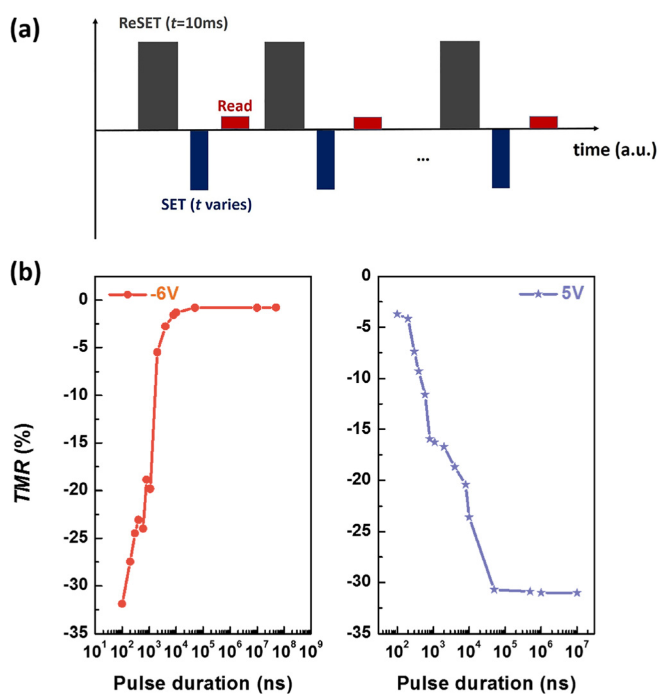


FIG. 3. (a) Schematic drawing of pulse trains used for control of TMR. A pulse train contains a ReSET pulse to switch the TMR into high or low reference state and then a SET pulse to drive the TMR into desired state, and eventually, the Read represents a TMR measurement. (b) The evolution of the TMR as a function of the writing pulse duration with amplitudes of −6 V and 5 V, respectively.

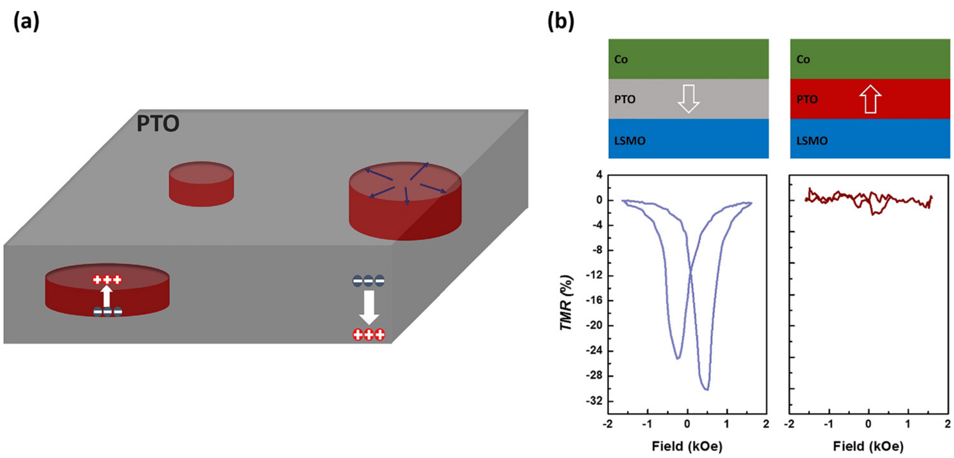


FIG. 4. (a) Schematic drawing of the multi-domain state consisting of oppositely polarised ferroelectric domains in the PTO tunnel barrier. The white arrows indicate the ferroelectric polarisation directions. (b) Typical TMR results for the Co/PTO/LSMO junction with PTO at fully up- and down-polarised states, respectively.

spin-filter so that the spin-polarisation of the current can strongly vary or even can change its sign while switching ferroelectric polarisation toward or away from ferroelectric/ferromagnetic electrode interfaces.[9–11,14,33] This behaviour is also confirmed in our work as shown in Fig. 4(b) where a clear TMR change by switching the PTO polarisation is presented.

Taking the multi-domain state and spin-dependent interfacial bonding effect at the ferroelectric/ferromagnetic metal interface into consideration, this multi-level TMR behaviour in our MFTJs can be qualitatively understood by a global TMR state resulting from a weighted arithmetic mean of different spin polarisations corresponding to mixed domains with opposite ferroelectric polarisation directions. In the ferroelectric tunnel memristor, the total device resistance is equivalent to individual resistors represented by ferroelectric domains connected in parallel.[26,28,29] Analogously, in the TMR case, each Co/PTO ferroelectric domain interface can control the spin polarisation and implicitly the TMR. The ferroelectric domains can be regarded as the separated spin transport channels where the spin polarisation is in the high state for polarisation pointing away from the PTO/Co interface and changes to its low state for the opposite situation when polarisation points to the PTO/Co interface, i.e., PTO domains are up-polarised. Within the top electrode area, the overall spin polarisation and consequently the TMR of the junction are therefore decided by the ratio of up- and down-polarised PTO domains.

In summary, we report a systematic study of multilevel control of TMR and TER in Co/PTO/LSMO MFTJs by using the ferroelectric domain configuration. A broad range of stable and reversible TMR and TER states between their own high and low states can be obtained via electric pulsing. This programmable TMR effect relies on the different spin-dependent properties between up- and down-polarised ferroelectric/ferromagnetic metal interfaces and stability of multi-domain state in ultrathin ferroelectrics. The interplay between electronic transport, magnetotransport properties, and ferroelectric domain configuration revealed in this study of these seemingly very simple MFTJ devices will further stimulate continued fundamental multiferroic research and broaden their applications.

*Note added in proof*: After this manuscript was submitted, the authors became aware of a very recent work reporting on magnetoelectrically coupled MFTJ memristors.[36]

See supplementary material for additional discussions about the (1) magnetic properties of the Co/PTO/LSMO structures, (2) retention properties about the MFTJ, and (3) PFM images of PTO ferroelectric domain configuration evolution by applying consecutive electric pulses.

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